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CHEMICAL REMEDIATION TREATMENTS ON PAH CONTAMINATED SOILS: CONSEQUENCES ON POLYCYCLIC AROMATIC COMPOUNDS (PAC)

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Polycyclic aromatic compounds (PAC), of which polycyclic aromatic hydrocarbons (PAH) are the most widely known, are common pollutants at contaminated sites (former gasworks, wood preservation facilities, coke oven plants). According to the European Environment Agency more than 13 % of all 1.5 million contaminated sites in Europe may house these pollutants. PAC-contaminated sites are of environmental concern because many PACs are known to be toxic, mutagenic and carcinogenic. However, during risk assessment of PAH-impacted sites, only a small subset of PAHs, often referred to as "the 16 priority PAHs", are generally monitored, in spite of the fact that many other contaminants also may be present. This may lead to a miscalculation of the risk posed by these sites, as well as of the detoxification result during remedial treatments of the sites.
The polar fractions of PAHs are often predominated by oxygenated PAHs (oxy-PAHs) and azaarens, but may also contain hydroxy-PAHs and nitro-PAHs for instance. The levels of oxy-PAHs and azaarenes are very often comparable to the PAH-levels, and sometimes they are even higher. In addition, some of these compounds, i.e. oxy-PAHs and hydroxy-PAHs may be formed as dead-end products during chemical and biological degradation of PAHs during remediation treatments as well as in natural attenuation context (especially with clay catalytic effect) and may thus potentially accumulate in remediation processes that are based on PAH-degradation. Many of the polar PACs have been highlighted as individual toxicants, mutagens and probable carcinogens, but the polar PACs are also of concern because of their relatively high solubility in water, which implies that they have higher tendency to spread via ground- and surface waters in the environment, as compared to the ordinary PAHs.. Despite these facts, semi-polar PACs are practically never included in the risk assessment of PAC-contaminated sites.
The evaluation of the efficiency of remediation processes (thermal desorption, ISCO, bioremediation...) is generally based on the measurement of specific parameters (Hydrocarbon index in the case of petroleum contamination, or 16 PAH US-EPA in the case of steel industries). Such limited characterisations do not allow evaluating the potential formation of organic by-products (especially PAC).
This work focus on identifying remediation processes that constitute the greatest risk or lowest risk in terms of polar PAC formation. We carried out test at laboratory scale selected remediation treatments on PAH contaminated soils in order to evaluate potential by-products formation (O-PAH especially).
Different remediation treatments, most commonly used and that may be the most problematic regarding O-PAH production, have been applied to three representatives PAH contaminated soils (former gasworks, coke oven plants and wood preservation facilities soils). These treatments include hydroperoxyde oxidation (H_2O_2), Fenton like oxidation using magnetite as catalyst, permanganate (MnO_4^-), and quicklime.
Each soil was sieved at 2mm, then freeze dried and crushed to 500µm. Such pre-treatment allows minimizing the heterogeneity. Initial soils exhibit contrasted general properties in term of total organic carbon, bitumen (organic fraction extractable in organic solvent) and PAC content.
Each remediation treatment has been carried out in batch condition during 1h, 24h and one week. Bitumen and PAH abatements were then measured by solvent extraction and GC-MS quantification. In parallel, degradation and/or formation of polar PAC during remediation experiment has been determined by GC-MS focusing especially on 12 O-PAC and 4 N-PAC.
These experiments enable the different remediation techniques to be compared with each other, and make it possible to estimate the purification efficiencies in relation to the pollution origin, the reaction kinetics and the PAH and polar PAC concentrations.

ThS C2.1 - Geohydrology & geostatistics

Wednesday | 17 April | 14.00-15.30 | Lecture hall 3

RISK ASSESSMENT OF CONTAMINATED SITES IN CLAYEY TILL SETTINGS: WILL EMBEDDED SAND BODIES PLAY A ROLE?

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Contaminated sites with chlorinated solvents are a major threat to groundwater. Risk assessment tools are needed in order to quantify leaching of contaminants into underlying aquifers. Low permeability settings such as clayey till pose a specific challenge because of fast vertical flow through fractures. Contamination migrating through the fractures spreads to the low permeability matrix by diffusion resulting in a long term secondary source of contamination due to back-diffusion. The clayey tills may also host geological strata with a large density of different types of sand bodies. In case these highly permeability, geological features form connected networks they could facilitate rapid spreading of contaminants in both lateral and longitudinal direction. However, these geological features have only been investigated sparsely, and their role for contaminant transport is basically unknown.
We present here the results of on-going research aiming for an improved understanding of contaminant transport and development of risk assessment tools in clayey till settings. The scope of the presentation will be: (1) to describe the presence and connectivity of sand bodies in clayey till settings, (2) to evaluate the contaminant transport in a clayey till setting with embedded sand bodies, (3) to assess the implications for risk assessment of contaminated sites.
A contaminated site (Vadsbyvej, Denmark) located in low-permeability till setting was used as a starting point to illustrate the role of geological heterogeneity on transport processes. In Vadsby, the clayey sediments comprise the upper 22-27 meter of the geological profile above a limestone aquifer. In the upper part macro pores and fractures were found to facilitate vertical migration of contaminants into greater depths. The clayey matrix is interspersed with sand lenses of fine and medium sands in a horizon between 10 to 15 m below ground surface.
The heterogeneity induced by sand lenses was characterized by means of outcrop studies at an analogue site located in 1.5 km distance from the Vadsbyvej site. On vertical outcrops the geometry and spatial variability of sand lenses were mapped and parameterized with use of geostatistical tools. Individual sand lenses have elongated and anisotropic shapes and vary in spatial extent. Most common features are sand pockets in the range of

1-3 m and smaller sand stringers with a thickness of only a few cm. We employed four different approaches to represent heterogeneity and hydraulic conductivity fields, respectively. The simplest method is to consider an equivalent porous medium, but with increasing amount of geological knowledge and conditioning data, stochastic simulations gave the best representation of sand lens distribution. Numerical solute transport modelling showed that the horizontal spreading of contaminants is significantly larger if the elongated, anisotropic shapes of sand lenses were well reproduced.
With regard to risk assessment at such sites, the location of monitoring wells is a major challenge. It was shown that depending on the spatial distribution of sand lenses, the contaminants may be diverted away already in the upper soil horizons escaping the monitoring wells installed beneath the source area. A fine scale representation of sand lenses and heterogeneity respectively is thus crucial for site investigation and proper risk assessment.
The general findings highlight a need for improved characterization of low permeability aquitards lying above aquifers used for water supply in order to perform proper risk assessment. The incorporation of a geological and hydrogeological framework should be encouraged as a part of future risk assessment.

GEOSTATISTICS FOR HIGHLIGHTING UNCERTAINTIES IN SOIL OR GROUNDWATER CONTAMINATION MANAGEMENT

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The management of a soil or groundwater contamination is based on an incomplete knowledge of the extension of the pollution and the concentration level. Indeed the concentration maps are built from drillings or wells in limited number.
The data exploratory analysis helps to delineate the effectively recognized domain and to detect the gaps of the survey. For example, the survey depth of polluted soils is often restricted to a few meters, which is often inadequate to reach the vertical extension of the pollution. The "source term" of the transfer to groundwater remains then inaccessible. Another type of difficulties is given for pesticide concentrations in groundwater by the "censored" large values (wells for drinkable water are deleted if the quality threshold is exceeded); and an important part of the low values is less than the quantification limit. In such cases, usual statistics can then not reflect reality and have to be adapted.
The measurements are used to establish concentration maps. From the same survey plan the precision of the estimation depends on the spatial variability of the studied substance. Furthermore the spatial (or temporal) variability increases generally with the concentration. This "proportional effect" have to be taken into account in order to deduce a realistic value for the standard deviation of the estimation error.
Going from the (estimated) concentration map to the demarcation of a polluted zone requires to take into account the estimation error, which is unknown but whose expectation and variance are modelled. Several non-linear estimation methods are used. The simplified conventional approach, based on a hypothesis of normal distribution of the estimation error, can supply non realistic bounds for a confidence interval. Indicator kriging or cokriging sometimes supplyes inconsistent results. More complex, disjunctive kriging (Matheron, 1973) is rarely used although it gives consistent results. If a spatial gaussian model is admissible on transformed data (which is easy to control) then the conditional probability is easy to calculate.
These techniques allow to establish a partition of the site in three zones with regard to a quality threshold, up to fixed statistical risks:

- The polluted supposed zone;
- The not polluted zone;
- The "zone of uncertainty", in which because of the estimation error, it is not possible to specify if the real concentration exceeds or not the threshold.

Examples are presented for cases of soil and groundwater contamination.
Finally, the question of the "support" for the comparison of the concentrations to a quality threshold is discussed.

CHARACTERIZATION OF A DEEP RADIOLOGICAL CONTAMINATION: INTEGRATION OF GEOSTATISTICAL PROCESSING AND HISTORICAL DATA

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Emerging in the early 80's, decommissioning is more than ever a major issue since hundreds of sites and facilities worldwide will end their operations over the next decades. Decontamination and remediation projects are all the more sensitive since they could last several years and turn out to be highly costly if not well-prepared. The key lies in an adequate contamination knowledge which helps to manage the remediation works and optimize the radiological waste production.
The methodologies commonly applied base their recommended decision-process on more or less complex statistical analyses to validate the remediation target after the waste removal work (guidance for demonstrating compliance with a dose- or risk-based regulation). These techniques ignore the spatial behavior of the contamination and the importance of sample localization upstream. The lack of representation and data processing tools leads to inefficient radiological characterizations, which always maximize the amount of contaminated soils or concrete volumes.
To solve these issues, the French Atomic Energy Authority (CEA) has developed a methodology over the last 10 years with Geovariances' partnership to fulfill the radwaste categorization. This methodology consists of an ordered sequence of evaluation actions starting with historical and functional analyses, in-situ characterization if relevant using non-intrusive measurement techniques, validation of contamination activity levels and depths with drill-holes and laboratory analyses. The implementation of geostatistics makes possible to give value to the collected data and map the contamination at each step of the sequence to finally get a robust and reliable characterization of contaminated areas. It also provides an efficient way out for sampling network optimization.
The communication introduces the geostatistical methodology and illustrates its added value on a real application case dealing with grounds of facilities partially dismantled at the end of the 1950s. 10 years ago, a first exploratory drill-hole confirmed the presence of a deep radiological contamination (more than 4m deep). More recently, 8 additional drill-holes failed to delineate the contaminated volumes according to a preliminary geostatistical analysis. The integration of the former topography and other geological data led to the realization of 7 supplementary drill-holes. This final stage significantly improved the characterization of the radiological contamination, which impacted the remediation project and the initially estimated volumes.
Finally, remediation work will be completed by the end of the year 2012 thus allowing us to get an industrial feedback by means of a comparison between predicted and excavated volumes.